

Challenges in Biomass–Thermoplastic Composites

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Abstract Wood and other biomass resources have been blended with thermoplastic such as polyethylene, polypropylene, polylactic acid and polyvinyl chloride to form wood plastic composites (WPC). WPCs have seen a large growth in the United States in recent years mainly in the residential decking market with the removal of CCA treated wood decking from residential markets. While there are many successes to report in WPCs, there are still some issues that need to be addressed before this technology will reach its full potential. This technology involves two different types of materials; one hygroscopic (biomass) and one hydrophobic (plastic) so there are issues of phase separation and compatibilization. This technology also involves two very different industries; wood and/or agricultural industries and plastic industries. Processing the two phases presents issues centered on maximizing mixing while minimizing damage to the biomass furnish. Melt flow index, processing temperatures, static electricity and density are also issues for the plastic industry which is used to high flowing high temperature processing conditions necessary when they either extrude plastics neat or use inorganic fillers such as glass, calcium carbonate or talc. While the biomass in the composite swells and shrinks due to moisture, the plastic phase swells and shrinks due to temperature. Freezing and thawing cycles also cause

problems as well as the effects of microorganism attack and ultraviolet radiation. Dyeing, refinishing, color stability and fasteners are also concerns. Solid extrusion, profile extrusion, co-extrusion, injection molding and thermal molding are all being used to make different WPC products in different parts of the world.

Keywords Wood · Thermoplastics · Moisture · Decay · Swelling · Ultraviolet degradation · Strength loss

Introduction

The use of lignocellulosics as fillers and reinforcements in thermoplastics has been gaining acceptance in commodity plastics applications in the past few years. It is interesting to point out that the use of the lignocellulosics in commodity thermoplastics to reduce cost and/or to improve mechanical performance is not new, and there are plenty of published papers, including patents dating back to the 1960's and 1970's. The resurgence of interest in the 1990's is probably due to increasing plastic costs and environmental aspects of using renewable materials.

The advantages of using a biobased component in thermoplastic composites is that the biobased resource is non abrasive, low in cost, widely available, sustainable, high filling levels possible, high specific properties, lower density per weight of raw material, flexible and recyclable [1]. The biobased resources that can be used in thermoplastics can be obtained from both forestry and agricultural resources. The physical form can vary from fine wood flour (particulate) to kenaf, flax, sisal, wood and other agro-based fibers [2]. The cost and performance of the final composite products dictates the type of lignocellulosic to be used in the plastics. The typical plastics used in this

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technology are various types of polyethylene, polypropylene, and polyvinyl chloride.

Biomass-fiber filled thermoplastics are being used in the auto industry, residential products, and many other applications with growing markets. Smith and Walcott [3] have pointed out four factors that are driving the WPC decking market: quality/cost ratio, impact of treating chemicals in the environment, builder acceptance, and effective marketing. There are, however, several major challenges that remain in this field that need to be solved including processing, use of additives, properties, and outdoor performance.

Processing

Typical blending of a biomass resource with a thermoplastic involves the plastic-filler/reinforcement to be shear mixed at temperatures above the softening point of the plastics. The heated mixture is then typically extruded into “small rods” that are then cut into short lengths to produce a conventional pellet. The pellets can then be used in typical injection or compression molding processes.

Processing the biomass component without loss of fiber properties is a major issue. Table 1 shows the loss of fiber length width and aspect ratio before and after processing oil palm fiber in a Brabender Plastic Corder with polypropylene at 180 °C. No matter what fiber length or width is used before processing, the fiber loses length and width during the processing. Many of the applications today use a biomass flour (aspect ration less than 10) rather than a fiber (aspect ratio greater than 10) so degradation of fiber length is not an issue. For using longer fibers, new technologies for mixing are needed to insure an even distribution of biomass in the thermoplastic matrix without damage to the fiber.

The processing temperatures used in the plastic industry to are usually above the decomposition temperature of the biomass materials. The plastic industry either does not use fillers or fillers such as glass, calcium carbonate or talc fillers so processing temperature can be very high to increase output. Temperatures above

180 °C result in decomposition of biomass so processing temperatures lower than 180 °C are needed if a biomass material is used as a filler. Thermoplastics with a high melt flow index and low softening temperatures are usually used.

Static electricity and density are also issues for the plastic industry that is used to high flowing high temperature processing conditions used when they either extrude plastics neat or use inorganics as fillers. The density of biomass materials is much lower than the inorganic fillers used and feeding an extruder can be a problem. Fibers are usually fed into the extruder in one of two ways. Pellets can be made of the biomass material and fed into the extruder along with the plastic pellets. A side stuffer can be added to the extruder line and the fiber or flour can be added at that point. If a loose fiber or flour is used, there is usually a build up of static electricity that has to be dealt with.

To reduce the cost of this blending process, direct injection molding can be done. The direct injection molding process probably has limitations on the amount of filler/fiber that can be used in the composite, and is also likely to be limited to particulate or shorter fiber. The chemical characteristics of the surface and bulk properties of the biomass are also important in the blending with plastics. The ability of the matrix of the lignocellulosic (hemicellulose and lignin) to soften at plastic processing temperatures may give these biomass materials unique characteristics to develop novel processing techniques.

Biomass fiber–thermoplastic composites can be produced by extrusion, injection molding, or thermoforming (compression molding). All three methods produce composites with different properties. Figure 1, for example, shows the moisture sorption of extruded, compression molded and injected molded wood flour-high density polyethylene (HDPE) as compared to the moisture sorption of solid wood [4]. The composites and wood were placed in a constant 65% relative humidity room (27 °C) and the weight gain measured as a function of time. Solid wood sorbs moisture the fastest and reaches equilibrium (9%) in about 10 days. Compression molded composites reach equilibrium (4.1%) in about 5 weeks, extruded reach

Table 1 Fiber length of oil palm before and after processing in a Brabender Plastic Corder with polypropylene at 180 °C

Average fiber length					
Before processing			After processing		
Length (mm)	Width (mm)	Aspect ratio	Length (mm)	Width (mm)	Aspect ratio
1.51	0.18	9.6	0.72	0.13	6.2
2.76	0.30	9.9	0.82	0.14	6.2
6.80	0.87	9.2	0.85	0.14	6.2

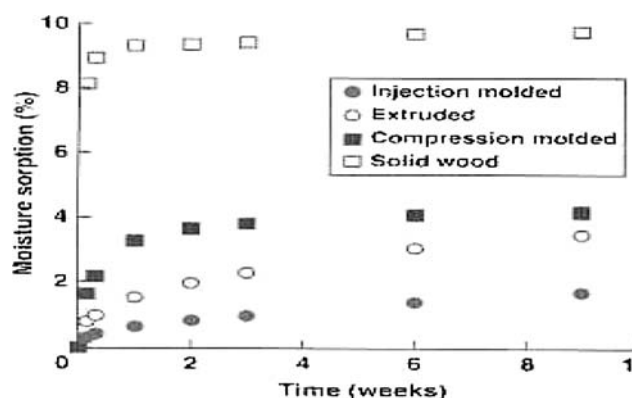


Fig. 1 Moisture sorption of high-density polyethylene containing 50% wood flour injected molded, extruded or compression molded and solid wood for comparison (Stark NM Personal Communication)

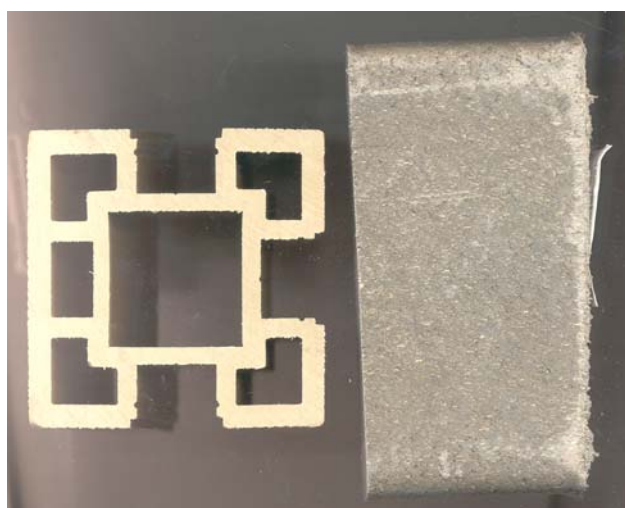


Fig. 2 Profile extrusion versus solid extrusion [4]

equilibrium (3.6%) in 9 weeks and injection molded had a moisture content of less than 2% and had not reached equilibrium at the end of the 9 week test. Moisture content as a function of time is an indication of the amount of biomass at or near the surface of the composite allowing moisture to be wicked into the interior. Injection molded composites have a surface rich in thermoplastic that surrounds and protects the biomass from coming into contact with moisture.

Figure 2 shows two different processing strategies for biomass–thermoplastic products: profile versus solid. More products can be made with the same quantity of material using profile extrusion and the products are lighter and can be stronger. Figure 3 shows an example of co-extrusion where a coating of vinyl is co-extruded over a wood fiber-thermoplastic core.

To reduce the weight and density of biomass–thermoplastic composites, foaming or blowing agents can be



Fig. 3 Co-extrusion of a fiber-thermoplastic core with a vinyl surface

dissolved or finely dispersed in the thermoplastic before processing [5]. A large number of bubble nuclei are generated and uniformly dispersed in the polymer matrix resulting in a light-weight composite when combined with a biomass fiber.

Equipment development is going forward to produce more efficient mixing systems that minimize fiber damage, faster processing through put, and new methods of product formation.

Use of Additives

Blending of the plastics with the biomass materials may require the use of many different types of additives. Compatibilizers to improve the dispersion, flow and mechanical properties of the composite are used when increased performance is required. Many of the applications of biomass materials in thermoplastics do not require a compatibilizer to improve mechanical properties but other chemicals may be added to improve UV stability and/or stability to heat or antimicrobials, antioxidants, colorants, or to improve impact strength. Processing aids or lubricants are almost always used. Foaming agents can also be added to reduce the density of the final product.

The most common compatibilizer used today is either maleic anhydride grafted polypropylene (MAPP) or maleic anhydride grafted polyethylene (MAPE). The mechanism of compatibilization is shown in Figs. 4 and 5. First, the anhydride end of the MAPP or MAPE reacts with a hydroxyl group on the surface of the biomass material

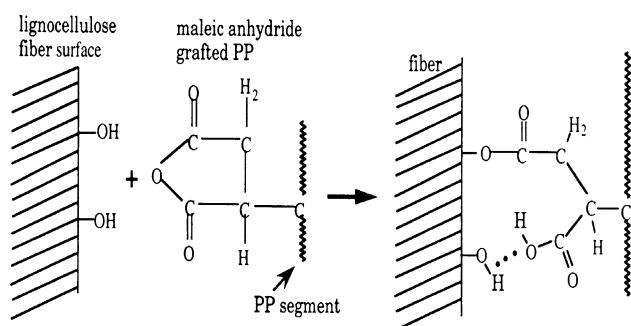


Fig. 4 Mechanism of MAPP reacting with a lignocellulosic surface hydroxyl group

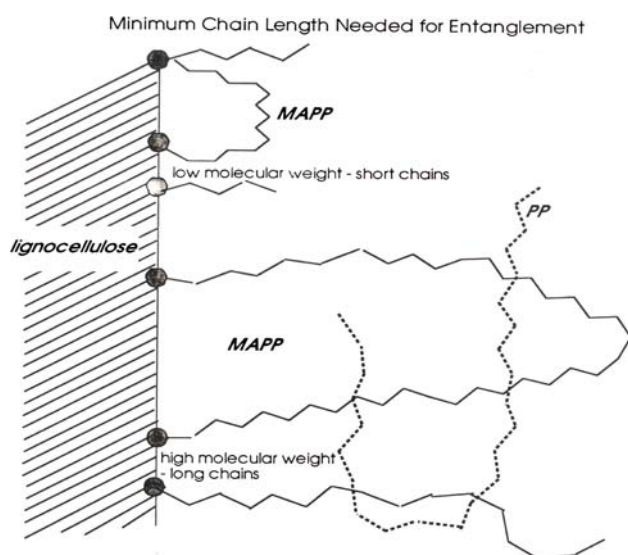


Fig. 5 Entanglement of melted polypropylene with the reacted MAPP/lignocellulosic

forming an ester linkage. The PP or PE tail on the grafted MAPP or MAPE then become entangled with the melted thermoplastic in the melt resulting in a mechanical link between the hydrophilic lignocellulosic and the hydrophobic thermoplastic.

Improvements in mechanical properties using a compatibilizer are shown in Fig. 6. The bottom curve in the stress–strain curve is PP alone. The dotted line is the curve for non-compatible PP with 50% kenaf loading and the next to the top curve is for compatibilized PP with 50% kenaf.

Colorants are commonly used both for aesthetics and UV protection. For a consistent color matching of the biomass materials and the thermoplastic, it may be necessary to add the colorant to both materials separately before extrusion. If the biomass fiber is very light in color and the thermoplastic dark, the composite will have light specks through out the composite.

Light stability in outdoor exposures is an area of considerable investigation [6, 7]. Most WPCs tend to lighten

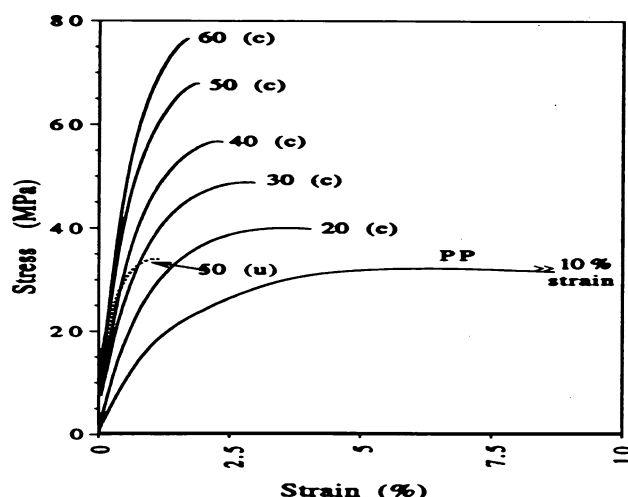


Fig. 6 Kenaf filled polypropylene at different filling levels with and without MAPP

over time [8]. Some manufacturers add pigments to slow this effect. Others add a gray pigment so that color change is less noticeable. Still others co-extrude a UV-stable plastic layer over the WPCs. In laboratory weatherometer tests, wood-PP composites lightened within 400–600 h of UV exposure. After 2,000 h of UV and water exposure, specimens were white and the white color continued throughout the 10 mm specimen. This in-depth color change may be due to stabilization of the free radicals formed from UV energy by the lignin in the wood allowing the free radicals to penetrate deeper into the wood structure [6].

The fire performance of WPC materials and products is just beginning to be investigated [9, 10]. These composites are different from many building materials in that they can melt as well as burn, making testing for fire resistance difficult.

Properties

The properties of biomass–thermoplastic composites depend on the volume fraction of biomass in the composite as well as processing temperature, additives, and type of thermoplastic used. Table 2 shows a comparison of mechanical properties of polypropylene filled with various materials [11]. The tensile and flexural properties of the kenaf-filled PP show significant improvement over those of unfilled PP. The tensile modulus of the kenaf-fiber system is higher than 40% calcium carbonate and talc filled PP, about equivalent to that of the 40%-mica PP but somewhat less than 40% glass filled PP. The properties of kenaf filled PP are especially favorable when compared in terms of specific properties since the biomass filler has a much lower density than the inorganic fillers. The notched Izod

Table 2 Comparison of mechanical properties of propylene with various fillers

Property	No filler	Kenaf	Talc	Calcium carbonate	Glass	Mica
% Filler by weight	0	50	40	40	40	40
% Filler by volume	0	39	18	18	19	18
Tensile modulus, GPa	1.7	8.5	4.0	3.5	9.0	7.6
Tensile strength, MPa	33	69	35	25	110	39
Flexural modulus, GPa	1.4	7.3	4.3	3.1	6.2	6.9
Flexural strength, MPa	41	98	63	48	131	62
Elongation at break, %	>10	2.2	–	–	2.5	2.3
Notched Izod impact, J/m	24	37	32	32	107	27
Water sorption, % in 24 h	0.02	1.05	0.02	0.02	0.06	0.03

impact strength of the kenaf filled PP is much lower than that of the glass filler PP but equivalent to talc, calcium carbonate and mica filled PP. Improvement in impact strength is a major challenge in biomass–thermoplastic composites.

Table 3 shows some mechanical properties of aspen fiber–polypropylene composites varying the volume fraction of the fiber with and without a compatibilizer (MAPP). Flexural and tensile strength and modulus increase with increasing fiber content. The fiber volume fraction has little effect on notched Izod but unnotched Izod goes down as the fiber content increases. The addition of the compatibilizer (2%) increases flexural and tensile strength and modulus but has little effect on notched Izod values.

Outdoor Performance

The proof of any material is in its performance. Most of the applications for fiber–thermoplastic composites have been for outdoor materials such as residential decking and

railing systems, windows, siding, roofing, door stiles, moldings, pallets, guard rails, sound barriers, curbing and furniture [3, 4].

Outdoor applications of biomass–thermoplastic composites require the materials to stand up to a wide variety of environmental factors including moisture, UV radiation, high temperatures, freeze-thaw cycles, mechanical action, and biological invasion. In almost all cases, more than one of these factors are acting at the same time and in cycles. A residential deck might be subject to all of these factors in a single year or less. Swelling, warping, twisting, mold growth, decay, color changes, and connector failure are some of the problems often encountered from normal outdoor use. For this reason, there has been a lot of environmental testing done both under laboratory conditions and outdoor in/above ground testing.

Table 4 shows the sorption of moisture as a function of time. Aspen fiber–polypropylene specimens with varying volume fractions of fiber with and without a compatibilizer (MAPP) were placed in a 90% RH room at 27 °C and the increase in weight measured over time. As the volume fraction of the fiber increases, the moisture sorption

Table 3 Mechanical properties of aspen fiber–polypropylene composites

Specimen	Flexural strength (MPa)	Flexural modulus (GPa)	Tensile strength (MPa)	Tensile modulus (GPa)	Izod notched (J/m)	Izod unnotched (J/m)
PP	27.9	1.38	26.2	1.69	22.4	713.5
PP/MAPP	34.6	1.79	29.3	1.82	18.6	563.3
30A/70PP	49.5	4.12	29.3	4.52	24.8	101.7
30A/68PP/MAPP	60.2	3.82	44.9	4.10	21.1	128.3
40A/60PP	54.6	4.60	34.9	5.22	19.6	85.5
40A/58PP/MAPP	66.4	4.66	47.7	5.14	19.8	108.7
50A/50PP	50.2	5.48	28.4	5.81	26.4	67.1
50A/48PP/MAPP	75.7	5.88	53.1	6.68	21.9	98.5
60A/40PP	45.9	6.09	25.6	6.95	23.9	55.2
60A/38PP/MAPP	75.8	6.73	48.1	7.19	21.3	81.1

Table 4 Moisture sorption of aspen fiber–polypropylene composites

Aspen/PP/MAPP	Weight gain (%)					
	25 Days	50 Days	75 Days	100 Days	150 Days	200 Days
0/100/0	0	0	0	0	0.2	0.4
30/70/0	0.7	1.4	1.7	2.1	2.4	2.8
30/68/2	0.7	0.7	1.1	1.5	1.5	2.2
40/60/0	0.7	1.4	1.7	2.0	2.4	3.0
40/58/2	0.4	1.2	1.5	1.9	2.7	3.5
50/50/0	1.3	2.0	2.6	3.6	4.3	5.3
50/48/2	1.5	1.8	2.2	2.9	4.0	5.1
60/40/0	3.7	4.5	5.6	6.0	6.3	6.7
60/38/2	1.6	2.2	3.5	4.4	5.1	6.0

increases and is still increasing after 200 days. There is a slight decrease in moisture sorption with the addition of MAPP indicating that the compatibilizer may allow the PP to come into closer contact with the fiber.

Figure 7 shows the weight change in an oven dry to water soaked test for 60% aspen fiber compounded with PP with and without MAPP. Each cycle represents 7 days of water soaking and one day of oven drying at 105 °C. The specimens without MAPP continue to gain weight after each water soaking cycle but there is less change in weight in the specimens containing MAPP.

Table 5 shows the influence of moisture on decay. Aspen fiber–polypropylene composites with various volume fractions of fiber were subjected to a 12 week ASTM standard soil block test using the brown-rot fungus *Gloeophyllum trabeum* or the white-rot fungus *Trametes versicolor*. Half of the specimens were placed in test directly from injection molding and half were soaked in water for 20 days before placing in test. The dry specimens were not degraded by either fungus but the water soaked specimens were attacked by both. The weight loss due to fungal attack increased with increasing fiber fraction. The compatibilizer in each fraction set did reduce the fungal attack.

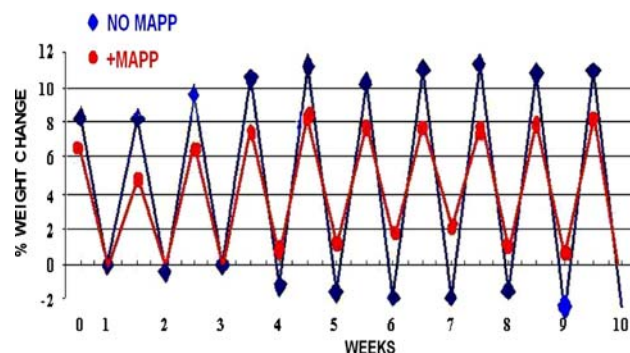


Fig. 7 Change in weight during a cyclic oven dried to water soaking of aspen fiber–polypropylene composites. ♦—60% aspen fiber, 40% PP ●—60% aspen fiber, 38% PP, 2% MAPP

Table 5 ASTM 12 week soil block test on dry and water soaked aspen fiber–polypropylene composites

Composition	Tested dry		Tested after water soaking	
	<i>G. trabeum</i> % wt. loss	<i>T. versicolor</i> % wt. loss	<i>G. trabeum</i> % wt. loss	<i>T. versicolor</i> % wt. loss
30/70	0.2	0	2.4	1.3
30/68/2	0.2	0	1.6	0.3
40/60	0.3	0	3.6	2.1
40/58/2	0.2	0	3.3	1.8
50/50	0.3	0	4.6	2.3
50/48/2	0.2	0	5.3	3.0
60/40	0.8	0	4.7	2.0
60/38/2	0.6	0	5.9	1.7

Figure 8 shows a railing in a state park in the Oregon made of biomass–thermoplastic material. Several types of micro-organisms are present growing on the surface of the composite. The decking and rails are also warped due to thermal expansion.

Figure 9 shows a badly warped deck in The Bahamas. The deck had also faded from its original dark green color due to UV exposure. Table 6 shows laboratory data on loss in flexural strength after 1,000 and 2,000 h of UV exposure (Stark NM Personal Communication). UV exposure results in both strength loss and color fading.

There is also research going on to improve connections/bonding between fiber–thermoplastic composite components, develop coating systems to change colors or improve the degraded aesthetics of the products, and to improve all aspects of performance in outdoor applications of biomass–thermoplastic materials.

Future Challenges

There is still a great deal of research and development going on in many parts of the world in the area of natural fiber/thermoplastic composites. Challenges in this



Fig. 8 Growth of several types of organisms on plastic lumber in an outdoor exposure in Oregon



Fig. 9 Warping due to thermal expansion of a plastic lumber deck in The Bahamas

Table 6 Loss of flexural strength due to UV exposure of a 50% wood flour 50% PE composite

Flexural strength		
Before exposure	After 1,000 h	After 2,000 h
40.2	37.9 (−2.3%)	31.2 (−9.0%)

research includes developing better mixing systems that minimize damage to the biomass fibers, developing better compatibilization systems, developing faster and

better extrusion/injecting/compression systems, improving impact strength, improving the properties of the biomass fraction through chemical, enzymatic and cold plasma modification to improve outdoor performance, advancing new profile and co-extrusions systems, developing more intensive testing procedures, and understanding performance through modeling.

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